412 Rec'd PCT/PTO 23 MAR 2000

(1390 REV. 5-93) US DEPT. OF COMMERCE PATENT & TRADEMARK OFFICE **TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371**		ATTORNEY'S DOCKET NUMBER 105034 U.S. APPLICATION NO. (if known, sec 37 C.F.R.1.5) 09/509121
INTERNATIONAL APPLICATION NO. PCT/JP99/03978	INTERNATIONAL FILING DATE July 23, 1999	PRIORITY DATE CLAIMED July 24, 1998

TITLE OF INVENTION
ELECTROLUMINESCENT DEVICE

APPLICANT(S) FOR DO/EO/US
Hidekazu KOBAYASHI

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A small entity statement.
16. ☐ Other items or information:

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17. ☒ The following fees are submitted:				CALCULATIONS	PTO USE ONLY
Basic National fee (37 CFR 1.492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO.....$840.00 International preliminary examination fee paid to USPTO (37 CFR1.482).....$670.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....$690.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....$970.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4).....$ 96.00					
ENTER APPROPRIATE BASIC FEE AMOUNT =				$840.00	
Surcharge of $130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	28- 20 =	8	X $ 18.00	$144.00	
Independent Claims	3- 3 =	0	X $ 78.00	$	
Multiple dependent claim(s)(if applicable)			+ $260.00	$	
TOTAL OF ABOVE CALCULATIONS =				$984.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28). -				$	
SUBTOTAL =				$984.00	
Processing fee of $130.00 for furnishing the English translation later than ☐ 20 ☐ 30 month from the earliest claimed priority date (37 CFR 1.492(f)). +				$	
TOTAL NATIONAL FEE =				$984.00	
				Amount to be refunded	$
				Charged	$

a. ☒ Check No. 107291 in the amount of $984.00 to cover the above fees is enclosed.
b. ☐ Please charge my Deposit Account No. _____ in the amount of $_____ to cover the above fees. A duplicate copy of this sheet is enclosed.
c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment, to Deposit Account No. 15-0461. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:
OLIFF & BERRIDGE, PLC
P.O. Box 19928
Alexandria, Virginia 22320

NAME: James A. Oliff
REGISTRATION NUMBER: 27,075

NAME: Thu A. Dang
REGISTRATION NUMBER: 41,544

(1390 Rev.8-93)

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ENTER APPROPRIATE BASIC FEE AMOUNT =				$840.00	
Surcharge of $130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	28- 20 =	8	X $ 18.00	$144.00	
Independent Claims	3- 3 =	0	X $ 78.00	$	
Multiple dependent claim(s)(if applicable)			+ $260.00	$	
TOTAL OF ABOVE CALCULATIONS =				$984.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28). -				$	
SUBTOTAL =				$984.00	
Processing fee of $130.00 for furnishing the English translation later than ☐ 20 ☐ 30 month from the earliest claimed priority date (37 CFR 1.492(f)). +				$	
TOTAL NATIONAL FEE =				$984.00	
				Amount to be refunded	$
				Charged	$

a. ☒ Check No. 107291 in the amount of $984.00 to cover the above fees is enclosed.
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(1390 Rev.8-93)

09/509121

410 Rec'd PCT/PTO 23 MAR 2000

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Hidekazu KOBAYASHI

Application No.: U.S. National Stage of PCT/JP99/03978

Filed: March 23, 2000 Docket No.: 105034

For: ELECTROLUMINESCENT DEVICE

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents
Washington, D. C. 20231

Sir:

Prior to initial examination, please amend the above-identified application as follows:

IN THE ABSTRACT:

Please substitute the attached Abstract for the Abstract currently in the application.

IN THE SPECIFICATION:

Please amend the specification as follows:

Page 1, line 1, delete "DECSRIPTION";

between lines 2 and 3, insert --BACKGROUND OF THE INVENTION--;

line 3, change "Technical Field" to --1. Field of the Invention--; and

line 6, change "Background Art" to --2. Description of Related Art--.

Page 2, line 9, change "Disclosure Of Invention" to --SUMMARY OF THE INVENTION--;

line 24, change "comprising" to --that may include--; and

line 25, change "composed of" to --including--.

Page 3, line 1, change "comprises" to --and may also include--;

line 18, after "composed of" insert --, for example,--; and

line 21, after "composed of" insert --, for example,--, and after "consisting of" insert --, for example,--.

Page 4, line 6, after "composed of" insert --, for example,--.

Page 5, line 7, after "composed of" insert --, for example,--;

line 19, change "comprising" to --that may include--;

line 20, after "composed of" insert --, for example,--; and

line 21, after "composed of" insert --, for example,--.

Page 6, line 1, change "Brief Description of the Drawings" to --BRIEF DESCRIPTION OF THE DRAWINGS--; and

line 14, change "Best Mode for Carrying Out the Invention" to --DESCRIPTION OF THE PREFERRED EMBODIMENTS--.

Page 7, line 2, after "composed of" insert --, for example,--;

line 9, after "composed of" insert --, for example,--; and

line 18, after "composed of" insert --, for example,--.

Page 8, line 18, after "composed of" insert --, for example,--.

Page 9, line 24, after "composed of" insert --, for example,--.

Page 10, line 6, after "composed of" insert --, for example,--.

Page 11, line 9, after "composed of" insert --, for example,--.

Page 13, line 4, after "composed of" insert --, for example,--.

Page 14, line 1, change "CLAIMS" to --WHAT IS CLAIMED IS:--.

IN THE CLAIMS:

Please cancel claims 1-14 without prejudice or disclaimer.

Please add claims 15-42 as follows:

--15. An electroluminescent device, comprising:

a light-emitting layer including at least an organic polymer and disposed between an anode and a cathode; and

a thin-film layer disposed at at least one of a position between the light-emitting layer and the anode, and a position between the light-emitting layer and the cathode, the thin-film layer suppressing unnecessary current that does not contribute to light emission.--

--16. The electroluminescent device according to claim 15, the thin-film layer being disposed only between the cathode and the light-emitting layer.--

--17. The electroluminescent device according to claim 15, the thin-film layer including at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal, a fluoride of an oxide of an alkaline earth metal, and a fluoride of an oxide of a group III element in the periodic table.--

--18. The electroluminescent device according to claim 16, the thin-film layer including at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal, a fluoride of an oxide of an alkaline earth metal, and a fluoride of an oxide of a group III element in the periodic table.--

--19. The electroluminescent device according to claim 15, the thin-film layer being disposed only between the anode and the light-emitting layer.--

--20. The electroluminescent device according to claim 15, further comprising:

a hole injection layer having electrical conductivity, the thickness thereof being not less than 100 nm, disposed between the light-emitting layer and the anode.--

--21. The electroluminescent device according to claim 15, further comprising:

a buffer layer having electrical conductivity, the thickness thereof being not less than 100 nm, disposed between the light-emitting layer and the anode.--

--22. The electroluminescent device according to claim 15, the organic polymer including at least one of polyfluorene and a derivative of polyfluorene.--

--23. The electroluminescent device according to claim 15, the organic polymer including at least one of poly(p-phenylenevinylene) and a derivative of poly(p-phenylenevinylene).--

--24. The electroluminescent device according to claim 15, the degree of polymerization of the organic polymer being at least two.--

--25. The electroluminescent device according to claim 15, the light-emitting layer being formed by depositing a plurality of light-emitting material layers.--

--26. The electroluminescent device according to claim 15, the light-emitting layer including the organic polymer being formed by a printing method.--

--27. The electroluminescent device according to claim 26, the printing method being an ink-jet method.--

--28. An electroluminescent device, comprising:

a light-emitting layer including at least an organic polymer and disposed between an anode and a cathode; and

a thin-film layer disposed at at least one of a position between the light-emitting layer and the anode, and a position between the light-emitting layer and the cathode, the thin-film layer suppressing unnecessary current that does not contribute to light emission, the organic polymer performing light emission in the wavelength range of 400 nm to 600 nm.--

--29. The electroluminescent device according to claim 28, the thin-film layer being disposed only between the cathode and the light-emitting layer.--

--30. The electroluminescent device according to claim 28, the thin-film layer including at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal, a fluoride of an oxide of an alkaline earth metal, and a fluoride of an oxide of a group III element in the periodic table.--

--31. The electroluminescent device according to claim 29, the thin-film layer including at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal, a fluoride of an oxide of an alkaline earth metal, and a fluoride of an oxide of a group III element in the periodic table.--

--32. The electroluminescent device according to claim 28, the thin-film layer being disposed only between the anode and the light-emitting layer.--

--33. The electroluminescent device according to claim 28, further comprising:

a hole injection layer having electrical conductivity, the thickness thereof being not less than 100 nm, disposed between the light-emitting layer and the anode.--

--34. The electroluminescent device according to claim 28, further comprising:

a buffer layer having electrical conductivity, the thickness thereof being not less than 100 nm, disposed between the light-emitting layer and the anode.--

--35. The electroluminescent device according to claim 28, the organic polymer including at least one of polyfluorene and a derivative of polyfluorene.--

--36. The electroluminescent device according to claim 28, the organic polymer including at least one of poly(p-phenylenevinylene) and a derivative of poly(p-phenylenevinylene).--

--37. The electroluminescent device according to claim 28, the degree of polymerization of the organic polymer being at least two.--

--38. The electroluminescent device according to claim 28, the light-emitting layer being formed by depositing a plurality of light-emitting material layers.--

--39. The electroluminescent device according to claim 28, the light-emitting layer including the organic polymer being formed by a printing method.--

--40. The electroluminescent device according to claim 28, the printing method being an ink-jet method.--

--41. An electroluminescent device, comprising:

a light-emitting layer including at least an organic polymer between an anode and a cathode; and

a layer including at least one of a fluoride of an alkali metal, a fluoride of an alkaline earth metal, and a fluoride of a group III element in the periodic table, the layer being disposed at at least one of a position between the light-emitting layer and the anode, and a position between the light-emitting layer and the cathode.--

--42. The electroluminescent device according to claim 41, the fluoride being lithium fluoride.--

REMARKS

Claims 15-42 are pending. By this Amendment, the specification and Abstract are amended to be placed in proper U.S. format and to correct minor informalities, claims 1-14

are canceled without prejudice or disclaimer, and claims 15-42 are added. Prompt and favorable examination on the merits is earnestly solicited.

Respectfully submitted,

James A. Oliff
Registration No. 27,075

Thu A. Dang
Registration No. 41,544

JAO:TAD/kmc

Attachment:
Substitute Abstract

OLIFF & BERRIDGE, PLC
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ABSTRACT OF THE DISCLOSURE

The invention provides an electroluminescent device having a structure that may include a light-emitting layer composed of, for example, at least an organic polymer between an anode and a cathode, and may also include a thin-film layer disposed between the light-emitting layer and the cathode, the thin-film layer suppressing unnecessary current which does not contribute to light emission.

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DESCRIPTION

ELECTROLUMINESCENT DEVICE

Technical Field

The present invention relates to structures of electroluminescent devices used for terminal components of information apparatuses, such as displays.

Background Art

Among recent aggressive activities for developing next generation luminous displays to replace cathode ray tubes (CRT) and liquid crystal displays, research and development of plasma display panels (PDP), field-emission displays (FED), organic electroluminescent displays have been actively performed. In organic electroluminescent materials, organic polymer materials emitting blue, green, and orange light, have been developed to such a level that the initial characteristics thereof can be utilized (The Society of Fiber Science and Technology, Japan, Symposium Abstracts 1998, 3A11, etc.). Polyfluorene derivatives have been well known as polymer materials for blue light emission, as described in Japanese Journal of Applied Physics, Vol. 30, No. 11B, November, 1991, pp. L1941 - L1943. In addition, as light-emitting materials having wavelengths of green or longer, poly(p-phenylenevinylene) derivatives have been well known, as described in USP 5,247,190.

Meanwhile, in electroluminescent devices using light-emitting materials having low molecular weights, it has been reported in Appl. Phys. Lett., 70, 152 (1997) that electron injection efficiency was improved by providing a cathode interface layer.

However, concerning organic polymer materials for blue light emission, even though the initial characteristics thereof can be satisfactory, there is the problem in that the wavelength of the luminescent color shifts toward the longer wavelength side with current-application time.

In addition, in electroluminescent devices using organic polymer materials as light-emitting materials, impurities exist therein due to difficulties in purifying organic polymers, and current which does not contribute to light emission flows through these impurities, whereby there is a problem in that sufficient efficiency cannot be obtained.

Furthermore, in a process for manufacturing electroluminescent devices, printing defects may occur when a printing method, specifically, an ink-jet method, is used as a method for forming a light-emitting layer, and electrical short circuits caused by the printing defects may occur, whereby there is a problem in that display may be disabled.

Disclosure of Invention

Taking the problems described above into consideration, the object of the present invention is to provide element configurations of electroluminescent devices using organic polymer materials, and more preferably, organic polymer materials for blue light emission as light-emitting materials, in which changes of luminescent color upon current application are suppressed and reliability is improved. In addition, the present invention provides element configurations capable of achieving satisfactory efficiency by suppressing unnecessary current, and provides element configurations capable of achieving satisfactory efficiency even for electroluminescent devices using organic polymer materials having wavelengths of green or longer. Furthermore, in a process for manufacturing display apparatuses by using a printing method, specifically, by using an ink-jet method, another object of the present invention is to prevent electrical short circuits at printing defects that may occur depending on the conditions.

According to the present invention, the electroluminescent devices described below are provided.

(1) An electroluminescent device having a structure comprising a light-emitting layer composed of at least an organic polymer and disposed between an anode and a cathode,

comprises a thin-film layer disposed at a position between the light-emitting layer and the anode, and/or between the light-emitting layer and the cathode. The thin-film layer suppresses unnecessary current which does not contribute to light emission.

According to the electroluminescent device described above, changes of luminescent color with current-application time can be effectively suppressed, and the reliability can therefore be noticeably improved. In addition, simultaneously, the insulating thin-film layer effectively blocks current passing through impurities existing in the organic polymer, and light-emitting efficiency is therefore improved.

Furthermore, in the present invention, configurations described below are provided as preferable embodiments.

(2) In the electroluminescent device described in (1) above, the organic polymer performs light emission in the wavelength range of 400 nm to 600 nm.

According to this configuration, an effect of improving the light-emitting efficiency described above can be obtained, specifically in the blue region and the vicinity thereof.

(3) In the electroluminescent device described in (1) or (2) above, the thin-film layer is disposed between the cathode and the light-emitting layer.

According to the configuration described above, unnecessary electron trap levels formed by joining the cathode and the light-emitting layer composed of the organic polymer at the interface thereof, can be avoided.

(4) In the electroluminescent device described in one of (1) to (3) above, the thin-film layer is composed of at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal; a fluoride of an oxide of an alkaline earth metal; and a fluoride of an oxide of a group III element in the periodic table.

According to the configuration described above, the thin-film layer can be readily formed by a deposition method, and due to the characteristics thereof, specifically,

changes of luminescent color with time are effectively suppressed, unnecessary current is suppressed, and light-emitting efficiency can therefore be improved.

(5) In the electroluminescent device described in (1) or (2) above, the thin-film layer is disposed between the anode and the light-emitting layer.

According to the configuration described above, hole trap levels formed by joining the anode and the light-emitting layer composed of the organic polymer can be avoided.

(6) In the electroluminescent device described in (1) or (2) above, a (positive) hole injection layer or a buffer layer having electrical conductivity, in which the thickness thereof is not less than 100 nm, is disposed between the light-emitting layer and the anode.

According to the configuration described above, changes of luminescent color with time after current-application are greatly and effectively decreased.

(7) In the electroluminescent device described in (1) or (2) above, the organic polymer is polyfluorene or a derivative thereof.

According to the configuration described above, the effect of the thin-film layer can be maximized specifically for blue light emission, and changes of luminescent color with time are greatly and effectively decreased.

(8) In the electroluminescent device described in (1) or (2) above, the organic polymer is poly(p-phenylenevinylene) or a derivative thereof.

According to the configuration described above, the light-emitting efficiency of the device for blue light emission can be noticeably improved.

(9) In the electroluminescent device described in (1) or (2) above, the degree of polymerization of the organic polymer is at least two.

According to the configuration described above, film formability of the light-emitting layer is improved, and improvements in reliability and characteristics are further achieved by disposing the thin-film layer.

(10) In the electroluminescent device described in (1) or (2) above, the light-emitting layer is formed by depositing a plurality of light-emitting material layers.

According to the configuration described above, an adjustable range for luminescent colors can be remarkably widened, and simultaneously, improvements in light-emitting efficiency and reliability can also be achieved.

(11) In the electroluminescent device described in (1) or (2) above, the light-emitting layer composed of the organic polymer is formed by a printing method.

According to the configuration described above, since the printing method, which is a very simple film-forming method, is used, and due to the provision of the thin-film layer, the probability of the occurrence of electrical short circuits is low even when printing defects exist, and display apparatuses having an exceedingly low probability of the occurrence of displaying defects can be obtained.

(12) In the electroluminescent device described in (11), the printing is an ink-jet method.

According to the configuration described above, the probability of the occurrence of electrical short circuits is low, even when printing defects caused by the ink-jet method exist, and display apparatuses having an exceedingly low probability of the occurrence of displaying defects can be manufactured.

In addition, as a specifically preferable embodiment, the present invention provides an electroluminescent device having a structure comprising a light-emitting layer composed of at least an organic polymer and disposed between an anode and a cathode, in which a layer composed of a fluoride of an alkali metal, an alkaline earth metal, or a group III element in the periodic table, is disposed between the light-emitting layer and the anode, and/or between the light-emitting layer and the cathode. In the devices described above, lithium fluoride is more preferably used as the fluoride.

Brief Description of the Drawings

Fig. 1 is a cross-sectional view of the structure of an electroluminescent device according to Example 1 of the present invention.

Fig. 2 shows a light emission spectrum of the electroluminescent device according to Example 1 of the present invention.

Fig. 3 shows a light emission spectrum of an electroluminescent device according to Comparative Example 1.

Fig. 4 shows a light emission spectrum of an electroluminescent device according to Example 2 of the present invention.

Fig. 5 shows the chromaticity of an electroluminescent device according to Example 4 of the present invention.

Fig. 6 is a cross-sectional view of the structure of an electroluminescent device according to Example 7 of the present invention.

Best Mode for Carrying Out the Invention

Preferable embodiments for carrying out the present invention will be described in detail with reference to the examples.

(Example 1)

In an electroluminescent device having a structure provided with an organic polymer and disposed between two electrodes (anode and cathode), one of which is transparent, according to this Example, an example will be described in which the organic polymer emits light in the wavelength range between 400 nm to 600 nm, and a thin-film layer is disposed between the organic polymer and the cathode.

In Fig. 1, a cross-sectional view of the electroluminescent device of the present invention is shown. As a transparent electrode (anode), an indium tin oxide (ITO) film was formed on a transparent glass substrate 1 and was then patterned. Next, as a (positive)

hole injection layer (transport layer) to be used as a thin-film layer 3, a 100-nm thick film composed of Bytron (Bayer AG) was formed by coating followed by drying thereof. Then, a xylene solution containing one percent of poly(dioctyl)fluorene was coated and dehydrated, and a 50-nm thick film thereof was obtained as a light-emitting layer 4. Subsequently, an ethyl acetate solution containing polymethylmethacrylate (PMMA) was coated and dehydrated, and a 5-nm thick film thereof was obtained as a thin-film layer 5. Calcium was deposited to a thickness of 100 nm as a cathode 6, and aluminum was then deposited to a thickness of 300 nm. Next, the unit thus formed was encapsulated by using a protective substrate and an encapsulating agent composed of ultraviolet-cured materials (ultraviolet curable epoxy resin).

The light emission spectrum of the light-emitting device (blue light-emitting device) thus prepared is shown in Fig. 2. The light-emitting efficiency thereof was 0.1 lm/W.

In this example, a polyfluorene derivative was used as the light-emitting layer 4; however, any material which emits blue light has the same effect.

After forming and patterning the ITO film, when formation of banks for isolating the cathodes and subsequent formation of layers thereon are performed, patterning of films composed of a cathode material is not specifically required. Meanwhile, without forming the banks thus mentioned, a cathode pattern may also be formed by patterning using a physical mask during cathode deposition.

When active elements, such as thin-film transistors (TFT), are formed on the glass substrate 1 beforehand, a large-scale displaying may be readily performed.

In this example, PMMA was used as the thin-film layer 5; however, an organic polymer having insulating properties, such as polyethylene, may be used in a manner similar to that described above. In addition, an inorganic material having insulating

properties, such as silicon dioxide, may be used as well. Concerning film formation, as well as a coating method, a deposition method or the like may also be employed.

In this example, ITO was used as the transparent electrode (anode); however, a transparent conductive material, such as IDIXO (Idemitsu K.K) and a NESA film, may also be used.

In this example, a glass substrate was used; however, a transparent substrate, such as a plastic, may also be used.

In this example, Bytron was used as the hole injection layer (transport layer) to be the thin-film layer 3; however, a material having electrical conductivity, such as polyaniline and a phthalocyanine compound, and an insulating material having a hole injection property, such as a phenylamine derivative of star-burst molecules, may also be used.

In this example, calcium was used as the cathode; however, a material having a small work function, such as lithium, magnesium, aluminum, and alloys thereof, may be used as well. Meanwhile, a material having a larger work function compared to that of a transparent electrode may be used by adjusting a driving voltage.

In this example, an ultraviolet cured material (ultraviolet curable epoxy resin) was used as the encapsulating agent; however, an encapsulating agent composed of a thermosetting resin having superior gas barrier properties and humidity resistance, may be used as well.

(Comparative Example 1)

An electroluminescent device without having the thin-film layer 5 (the thin-film layer between the light-emitting layer and the cathode) of the structure shown in Fig. 1 of Example 1, was prepared. The light emission spectrum thereof is shown in Fig. 3. The light-emitting efficiency was 0.06 lm/W.

(Example 2)

In this Example, an example will be described, in which a fluoride of an oxide of an alkali metal; a fluoride of an oxide of an alkaline earth metal; or a fluoride of an oxide of a group III element in the periodic table, is used for the thin-film layer of the structure shown in Fig. 1.

Formation methods for layers other than the thin-film layer were the same as described in Example 1. As the thin-film layer 5, calcium fluoride film formed by deposition to a thickness of 2 nm was used. The light emission spectrum of the light-emitting device (blue light-emitting device) thus prepared is shown in Fig. 4. The light-emitting efficiency was 0.17 lm/W.

In this example, calcium fluoride was used by deposition as the thin-film layer; however, lithium fluoride may also be used. In addition, a fluoride of an oxide of an alkali metal, such as lithium, sodium, or potassium; a fluoride of an oxide of an alkaline earth metal, such as beryllium, magnesium, calcium, or scandium; and a fluoride of an oxide of a group III element in the periodic table, such as boron, aluminum, or gallium, may also be used. In addition, a material that has adequate electrical insulating properties, easy film formability, and suppression of unnecessary current which does not contribute to light emission, may also be used.

(Example 3)

In this Example, an example will be described in which poly(p-phenylenevinylene) or a derivative thereof is used as an organic polymer for the light-emitting layer. The structure of the Example other than an organic polymer layer (light-emitting layer) was equivalent to that of the light-emitting device described in Example 1.

As the light-emitting layer 4 (layer composed of an organic polymer) in Fig. 1, precursors of poly(p-phenylenevinylene) were coated and baked, and a 100-nm thick film

was thereby obtained.

The light-emitting efficiency of the electroluminescent device thus prepared was 1.16 lm/W.

(Comparative Example 2)

In Comparative Example 1, when a poly(p-phenylenevinylene) film was formed and used as the light-emitting layer (layer composed of an organic polymer) in a manner similar to that of Example 3, the light-emitting efficiency thereof was 0.4 lm/W.

(Example 4)

In this Example, in the light-emitting device having the structure (Example 1) shown in Fig. 1, an example will be described in which a hole injection layer or a buffer layer having electrical conductivity, which is disposed between the light-emitting layer 4 and the anode 2 as the thin-film layer 3, was formed at various thickness.

In Example 1, the electroluminescent devices were prepared by changing the thickness of the hole injection layers from 25 nm to 220 nm, and the chromaticities of these electroluminescent devices measured five minutes after current-application are shown in Fig. 5. It was clear that chromaticity shifting to a blue side could be obtained concomitant with an increase in the thickness of the buffer layer (specifically, not less than 100 nm).

(Example 5)

In this Example, in the light-emitting device having the structure (Example 1) shown in Fig. 1, an example will be described in which the degree of polymerization of the organic polymer was changed. When the degree of polymerization was changed to 1, 2, and 1,000, film formability of an organic polymer having the degree of polymerization of 1 was seriously inferior. Meanwhile, concomitant with an increase of the degree of polymerization, superior film formability was obtained and an effect by inserting the thin-

film layer was enhanced. Even when the degree of polymerization was 2, the effect by disposing the thin-film layer could be observed.

(Example 6)

In this Example, an example, in which an ink-jet method was employed for forming the light-emitting layer, will be described.

Forming methods other than the formation of the light-emitting layer were equivalent to those described in Example 2. The formation of the light-emitting layer was performed by the ink-jet method. At the pixel on which the light-emitting layer could not be coated for some reason, a stacked layered structure composed of an ITO/a hole injection layer (transport layer) (Bytron of Bayer AG in this Example)/a thin-film layer (2 nm of LiF in this Example)/Ca/Al was formed. The measured current density of this structure was not greater than 1 mA/cm^2, and when the light-emitting layer was formed, the current density was a few tens of mA/cm^2. It is understood that current was suppressed when the light-emitting layer was not formed.

In this Example, the ink-jet method was described; however, other printing methods may also be used.

(Example 7)

In this Example, an example in which two light-emitting layers were formed will be described. In Fig. 6, the structure of an electroluminescent device of this Example is shown.

An anode group 52 was formed on a substrate 51, and subsequently, a bank 53 and a hole injection layer (transport layer) 54 (20-nm thick Bytron film by Bayer AG in this Example) 54 were formed. Next, on a pixel for red light emission, a solution containing precursors of poly(p-phenylenevinylene) doped with one percent of rhodamine 101 (RPPV) was coated as a first light-emitting layer (55) by an ink-jet method and then baked

at 150°C for 4 hours in N_2 atmosphere, and a 40-nm thick film was thereby obtained. Then, on a pixel for green light emission, a solution containing precursors of poly(p-phenylenevinylene) (PPV) was coated as a second light-emitting layer (55') by an ink-jet method and was then baked at 150°C for 4 hours in N_2 atmosphere, and a 30-nm thick film was thereby obtained. On a pixel for blue light emission, nothing was coated by an ink-jet method. Subsequently, a xylene solution containing poly(dioctyl)fluorene was spin-coated on the entire pixels to a thickness of 45 nm as a third light-emitting layer (56). Next, a 2-nm thick film of lithium fluoride was formed by deposition over the entire surface of the substrate as a thin-film layer 57, and then a calcium film and an aluminum film were formed as a cathode 58 by deposition to a thickness of 100 nm and 200 nm, respectively. Over the unit thus prepared, a protective layer 59 was formed by a protective substrate and an encapsulating material. In addition, lead electrodes were connected to a controller circuit, whereby display was performed.

In the electroluminescent device thus prepared, the efficiency of the red light-emitting pixels was 0.15 lm/W, the efficiency of the green light-emitting pixels was 0.12 lm/W, and the efficiency of the blue light-emitting pixels was 0.18 lm/W.

A display panel (number of pixels was 320 × 240 and size was 2 inch) was made by forming a TFT device in each pixel of the substrate 51 beforehand. Electrical consumption was approximately 1.6 W and display luminance was 30 Cd/m^2, when an active matrix driving method displayed animation.

The thickness of each layer described in this Example is not limited thereto. In addition, the light-emitting materials are not limited to those described in this Example. When TFT arrays are formed on the substrate to be used, animation can be displayed. Meanwhile, when anodes and cathodes are formed as electrode groups in the form of stripes, and are disposed to orthogonally cross each other, simple matrix driving can be

performed.

As described above in detail, according to the present invention, by disposing a thin-film layer, which suppresses unnecessary current which does not contribute to light emission, between a light-emitting layer composed of an organic polymer and a cathode, a tendency of the luminescent color to shift to longer wavelengths can be suppressed, and a light-emitting efficiency can be noticeably improved. In addition, even when defects of the light-emitting layer occur during a forming process therefor using a printing method or the like, electrical short circuits can be effectively prevented. Consequently, an organic electroluminescent (EL) display, which has consistent performance, a high light-emitting efficiency, and high color reproducibility, can be simply produced and can be provided, and the application thereof to information display apparatuses will therefore be accelerated.

Industrial Applicability

The electroluminescent device according to the present invention can be suitably applied to electronic devices, such as lap-top type personal computers (PC) which are required to display high quality images, televisions, viewfinder type or direct-view monitor type video tape recorders, car navigation devices, electronic notebooks, electronic calculators, word processors, engineering work stations (EWS), mobile phones, video telephones, POS terminals, pagers, and apparatuses provided with touch panels.

CLAIMS

(1) An electroluminescent device having a structure comprising a light-emitting layer composed of at least an organic polymer and disposed between an anode and a cathode, wherein the electroluminescent device comprises a thin-film layer disposed at least at a position between the light-emitting layer and the anode, and between the light-emitting layer and the cathode, the thin-film layer suppressing unnecessary current which does not contribute to light emission.

(2) An electroluminescent device according to Claim 1, wherein the organic polymer performs light emission in the wavelength range of 400 nm to 600 nm.

(3) An electroluminescent device according to Claim 1 or Claim 2, wherein the thin-film layer is disposed between the cathode and the light-emitting layer.

(4) An electroluminescent device according to one of Claims 1, 2, and 3, wherein the thin-film layer is composed of at least one material selected from the group consisting of a fluoride of an oxide of an alkali metal; a fluoride of an oxide of an alkaline earth metal; and a fluoride of an oxide of a group III element in the periodic table.

(5) An electroluminescent device according to Claim 1 or Claim 2, wherein the thin-film layer is disposed between the anode and the light-emitting layer.

(6) An electroluminescent device according to Claim 1 or Claim 2, further comprising a hole injection layer or a buffer layer having electrical conductivity, the thickness thereof being not less than 100 nm, disposed between the light-emitting layer and the anode.

(7) An electroluminescent device according to Claim 1 or Claim 2, wherein the organic polymer comprises polyfluorene or a derivative thereof.

(8) An electroluminescent device according to Claim 1 or Claim 2, wherein the organic polymer comprises poly(p-phenylenevinylene) or a derivative thereof.

(9) An electroluminescent device according to Claim 1 or Claim 2, wherein the degree of

polymerization of the organic polymer is at least two.

(10) An electroluminescent device according to Claim 1 or Claim 2, wherein the light-emitting layer is formed by depositing a plurality of light-emitting material layers.

(11) An electroluminescent device according to Claim 1 or Claim 2, wherein the light-emitting layer composed of the organic polymer is formed by a printing method.

(12) An electroluminescent device according to Claim 11, wherein the printing method is an ink-jet method.

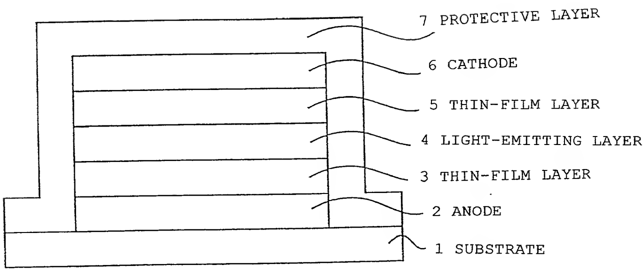
(13) An electroluminescent device having a structure comprising a light-emitting layer composed of at least an organic polymer between an anode and a cathode, wherein the electroluminescent device comprises a layer composed of a fluoride of an alkali metal, an alkaline earth metal, or a group III element in the periodic table, the layer being disposed at least at a position between the light-emitting layer and the anode, and between the light-emitting layer and the cathode.

(14) An electroluminescent device according to Claim 13, wherein the fluoride is lithium fluoride.

ABSTRACT

An electroluminescent device having a structure comprising a light-emitting layer (4) composed of at least an organic polymer between an anode (2) and a cathode (6), comprises a thin-film layer (5) disposed between the light-emitting layer (4) and the cathode (6), the thin-film layer suppressing unnecessary current which does not contribute to light emission.

FIG. 1


7 PROTECTIVE LAYER
6 CATHODE
5 THIN-FILM LAYER
4 LIGHT-EMITTING LAYER
3 THIN-FILM LAYER
2 ANODE
1 SUBSTRATE

09/509121

FIG. 2

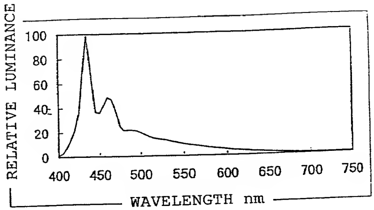


FIG. 3

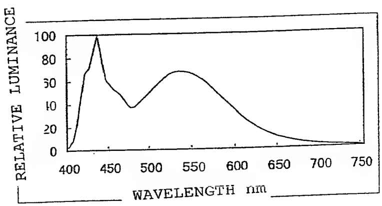


FIG. 4

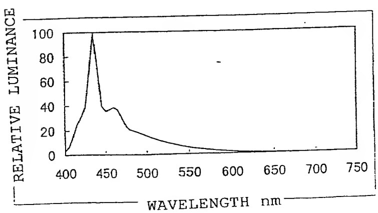


FIG. 5

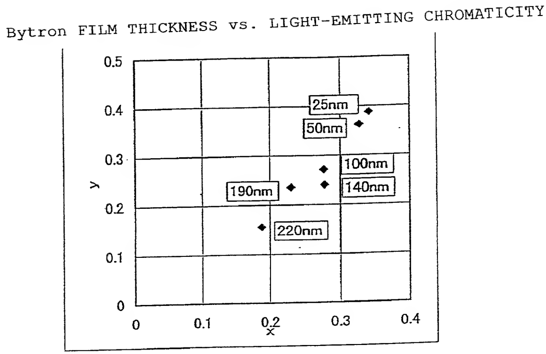
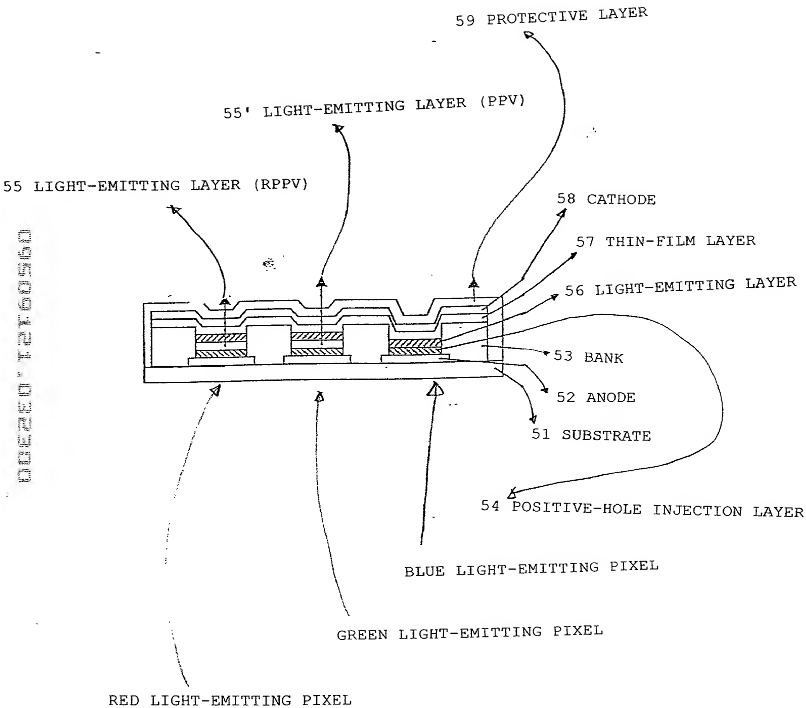

Bytron FILM THICKNESS vs. LIGHT-EMITTING CHROMATICITY
0.5
0.4
0.3
0.2
0.1
0
y
0
0.1
0.2
0.3
0.4
x
25nm
50nm
100nm
140nm
190nm
220nm

FIG. 6


59 PROTECTIVE LAYER
55' LIGHT-EMITTING LAYER (PPV)
55 LIGHT-EMITTING LAYER (RPPV)
58 CATHODE
57 THIN-FILM LAYER
56 LIGHT-EMITTING LAYER
53 BANK
52 ANODE
51 SUBSTRATE
54 POSITIVE-HOLE INJECTION LAYER
BLUE LIGHT-EMITTING PIXEL
GREEN LIGHT-EMITTING PIXEL
RED LIGHT-EMITTING PIXEL

PTO/SB/106 (8-96)
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Seiko Epson Ref. No.: F004635US00 Attorney's Ref. No.: 105034

Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は、下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

電界発光素子

ELECTROLUMINESCENT DEVICE

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the specification of which is attached hereto unless the following box is checked:

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☐ was filed on ＿＿ as United States Application Number or PCT International Application Number ＿＿ and was amended on ＿＿ (if applicable).

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I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

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I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

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I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)
外国での先行出願

(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	Priority Not Claimed 優先権主張なし
10-210012	Japan	24/July/1998	☐
11-203632	Japan	16/July/1999	☐

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(Application No.) (出願番号)	(Filing Date) (出願日)	(Application No.) (出願番号)	(Filing Date) (出願日)

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(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況：特許許可済、係属中、放棄済)
PCT/JP99/03978	23/July/1999	Pending

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

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国籍	Citizenship
私書箱	Post Office Address
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